Sources, Seasonality, and Trends of Southeast US Aerosol: An Integrated Analysis of Surface, Aircraft, and Satellite Observations with the GEOS-Chem Chemical Transport Model

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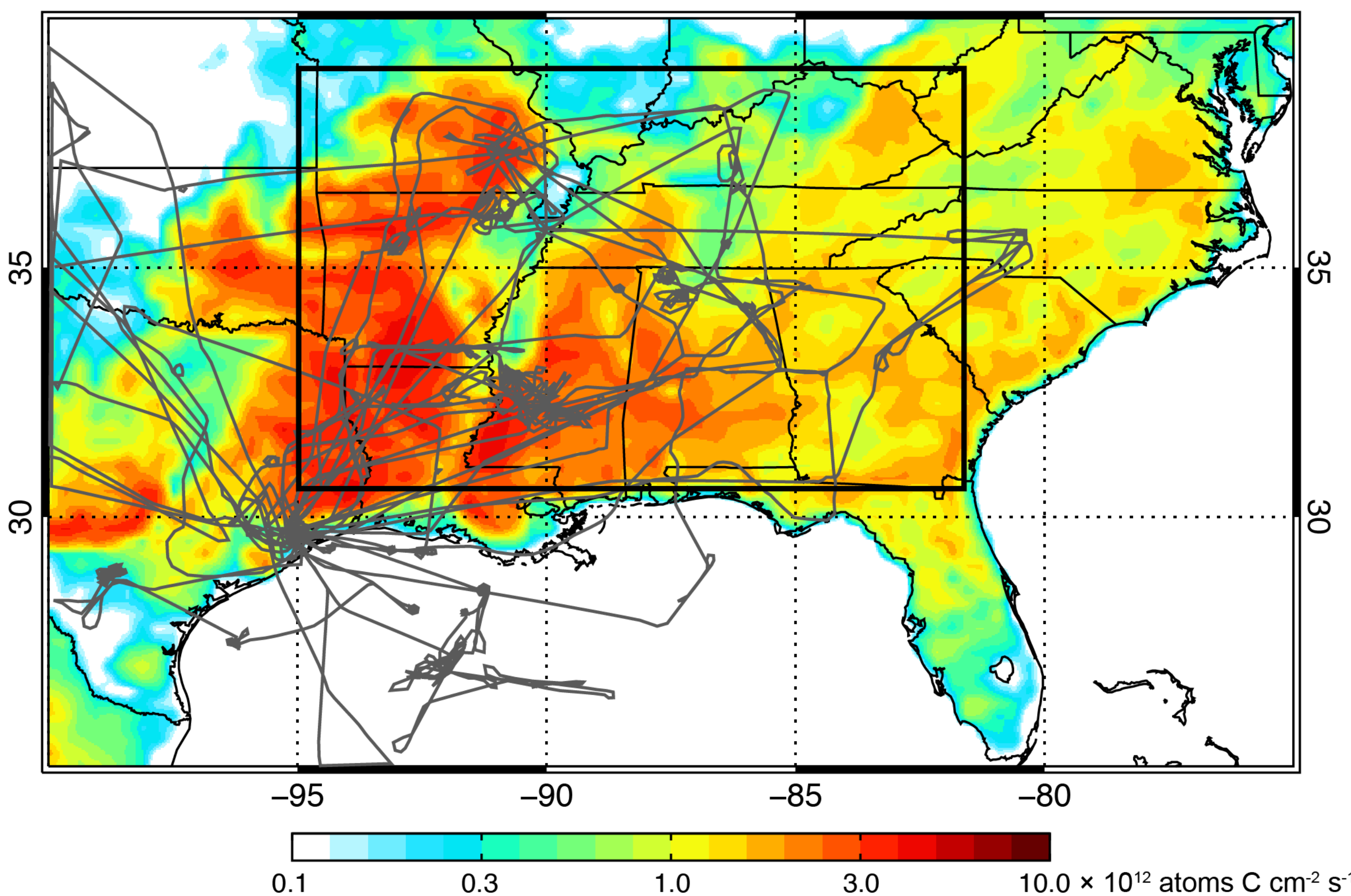
SEAC^{4}RS Flight Tracks and MEGAN2.1 Isoprene Emissions



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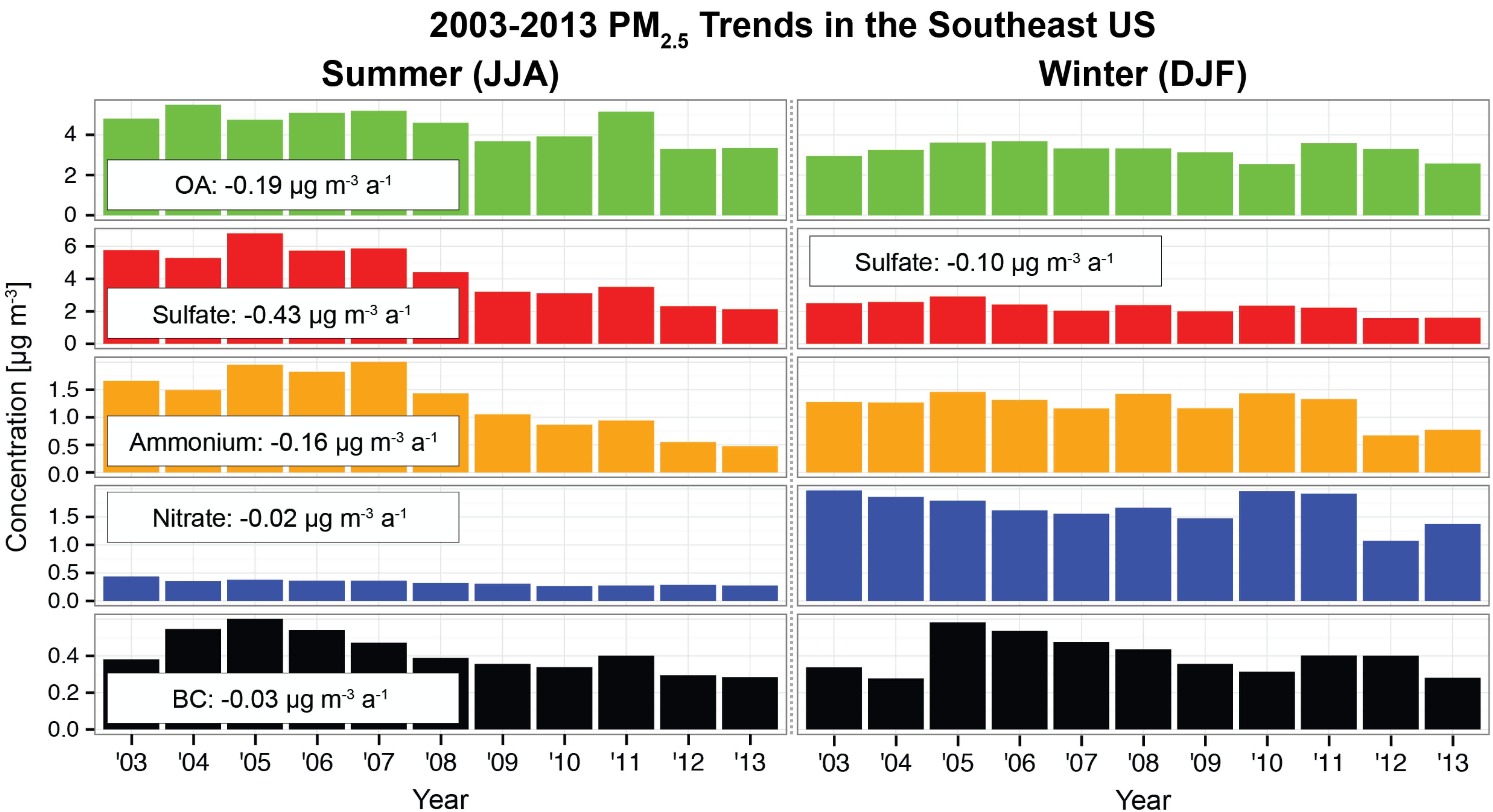
ATMOSPHERIC CHEMISTRY MODELING GROUP

Studies of Emissions & Atmospheric Composition, Clouds & Climate Coupling by Regional Surveys
NASA - NRL
SEAC4RS
Earth Science 2013

Air Quality in the Southeast US is Rapidly Changing

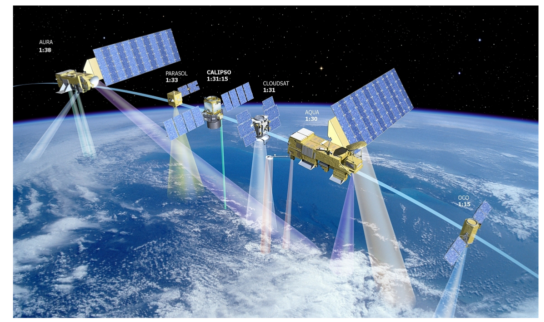


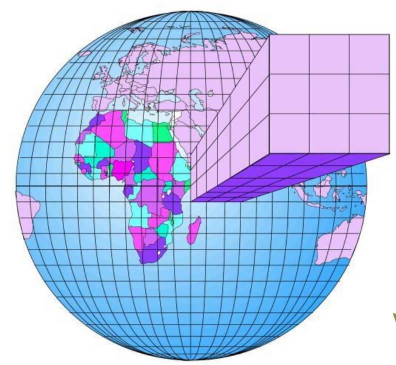
- The change in the relative importance of OA and sulfate has implications for aerosol hygroscopicity, light extinction, and radiative forcing
- Ammonium concentrations have declined at the same rate as sulfate

Analysis Framework

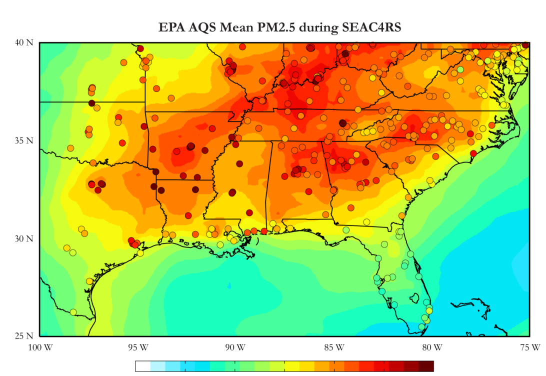
Satellites

MODIS, MISR





GEOS-Chem
as a transfer function



Surface Sites

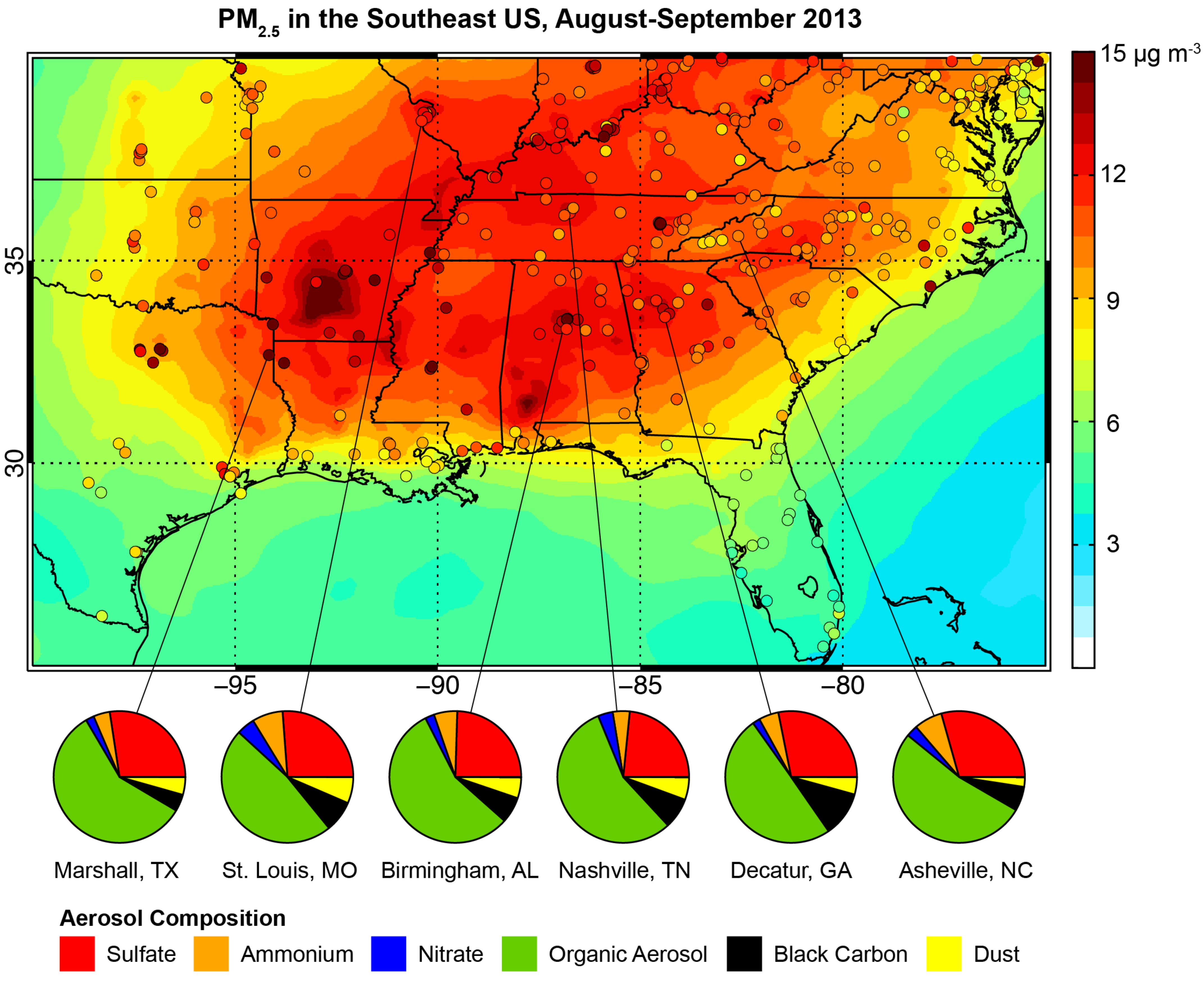
CSN, IMPROVE, SEARCH, AERONET

Aircraft Data

NASA SEAC⁴RS Campaign

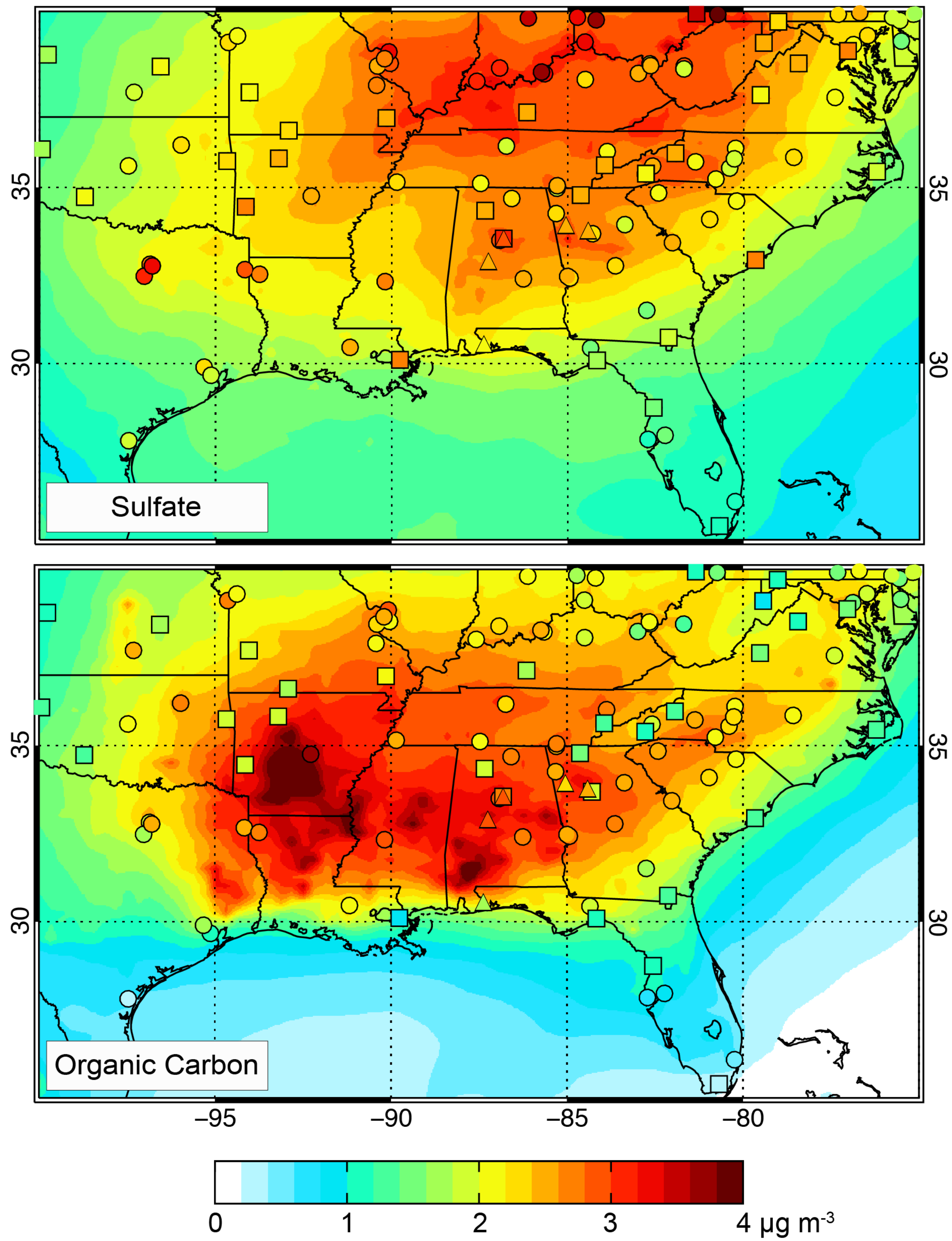
1. Determine the consistency between different sets of measurements
2. Interpret the measurements in terms of their implications for the sources of sulfate and OA in the Southeast US
3. Explain the seasonal aerosol cycle in the satellite and surface data
4. Assess the ability of CTMs to relate satellite measurements of AOD to surface PM

$PM_{2.5}$ Composition and Distribution is Fairly Homogeneous

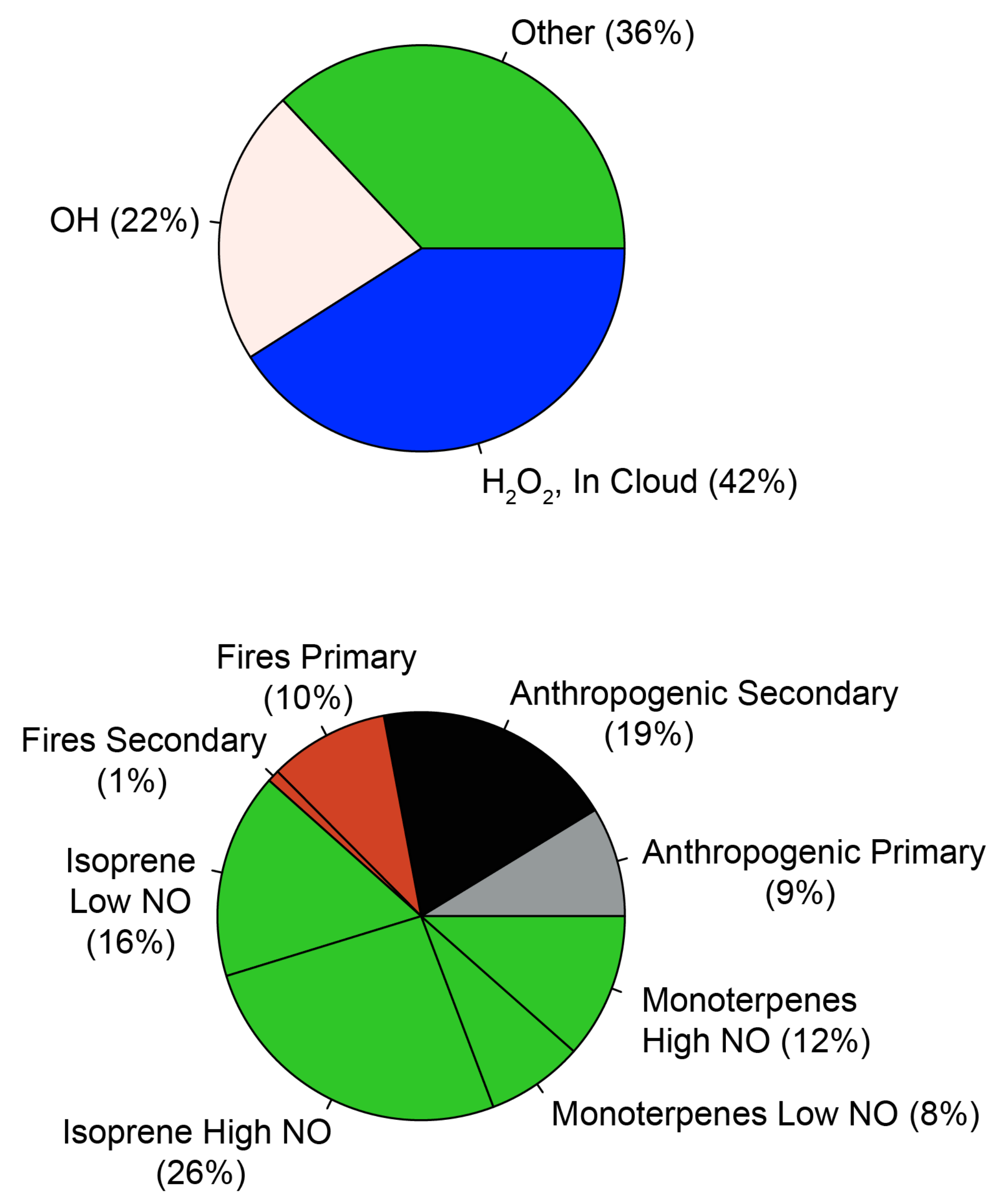


OC is Predominantly from Biogenic Sources, Mostly Isoprene

Mean Sulfate and OC, August-September 2013

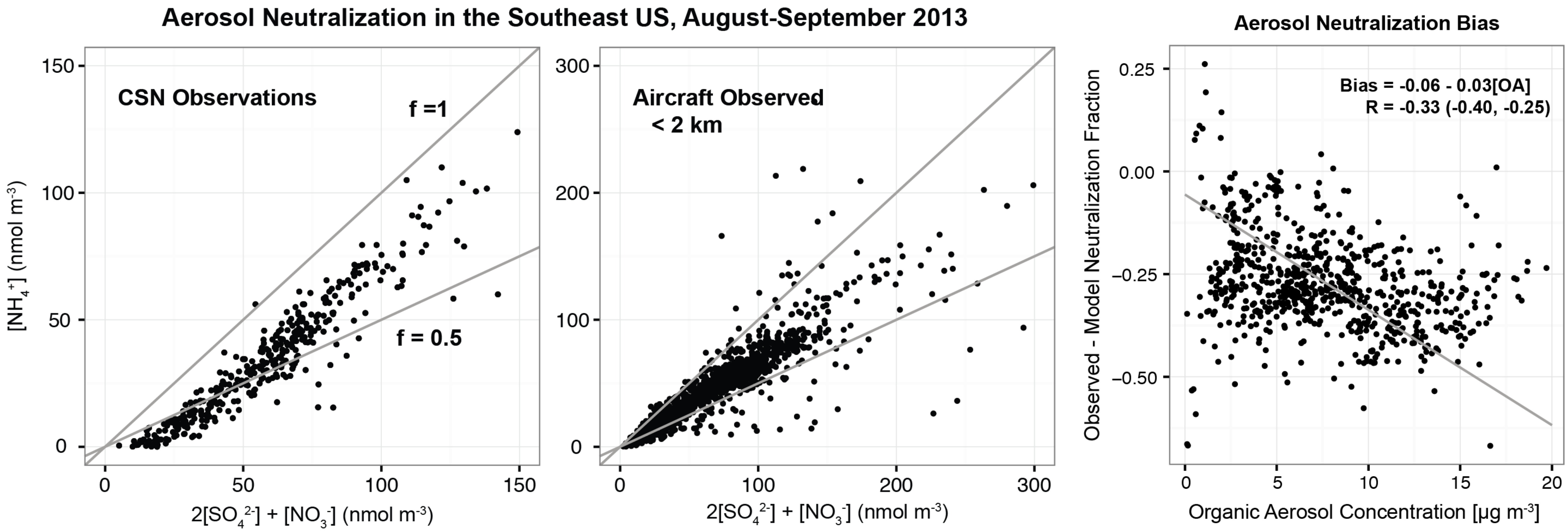


Source Attribution



Successful simulation of sulfate requires a missing oxidant - Criegees, aqueous aerosols, clouds?

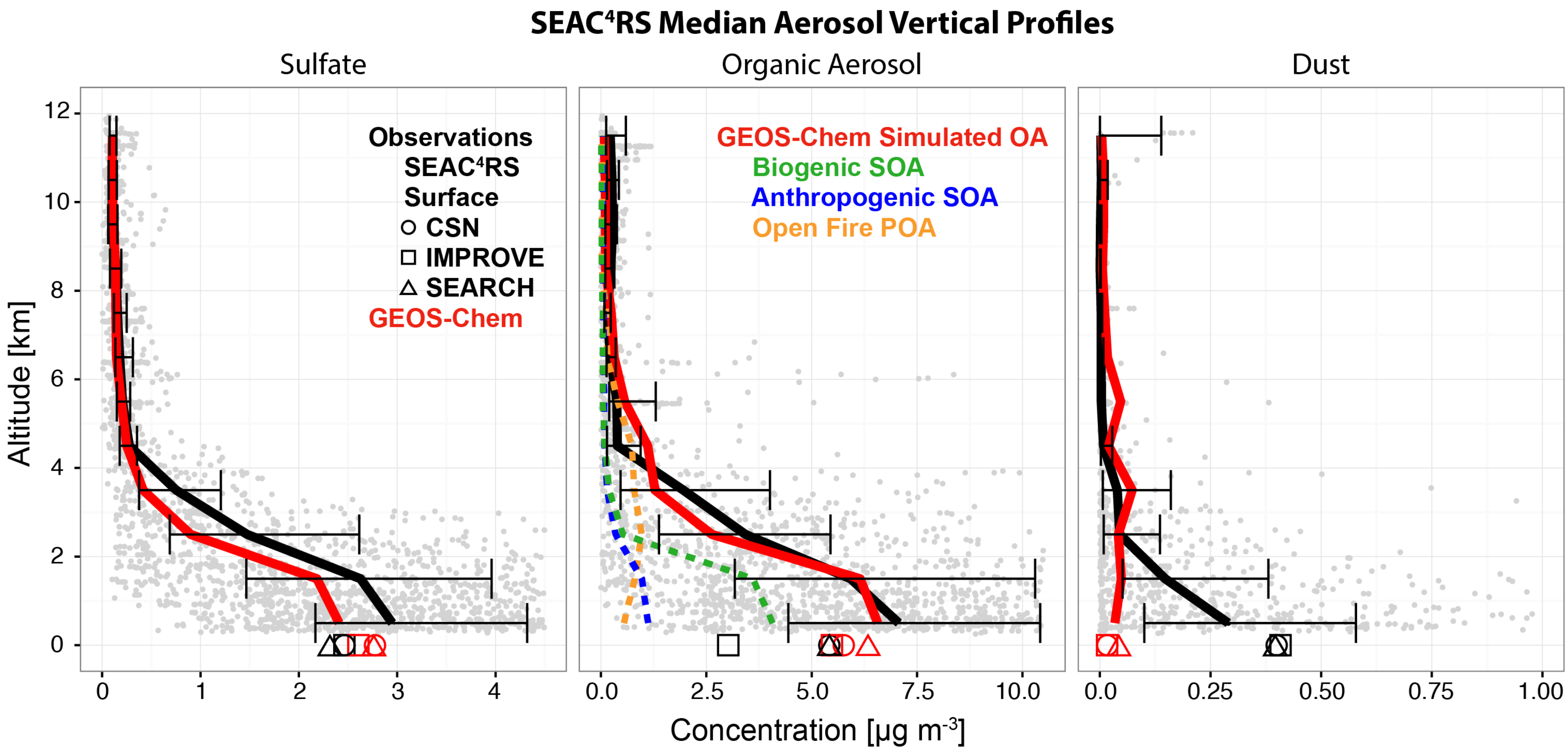
Understanding the Ammonium Trend in the Southeast US



- No evidence for decline in ammonia emissions from wet deposition data
- Wet deposition data also shows ammonium is in excess of sulfate
- Ammonium trend is inconsistent with an extent of neutralization < 1 in surface aerosol, unless there is inhibition of ammonia uptake. Could this be from organic particle material?
- This will have implications for aerosol phase, hygroscopicity, and formation of aerosol nitrate

AMS: Jose Jimenez

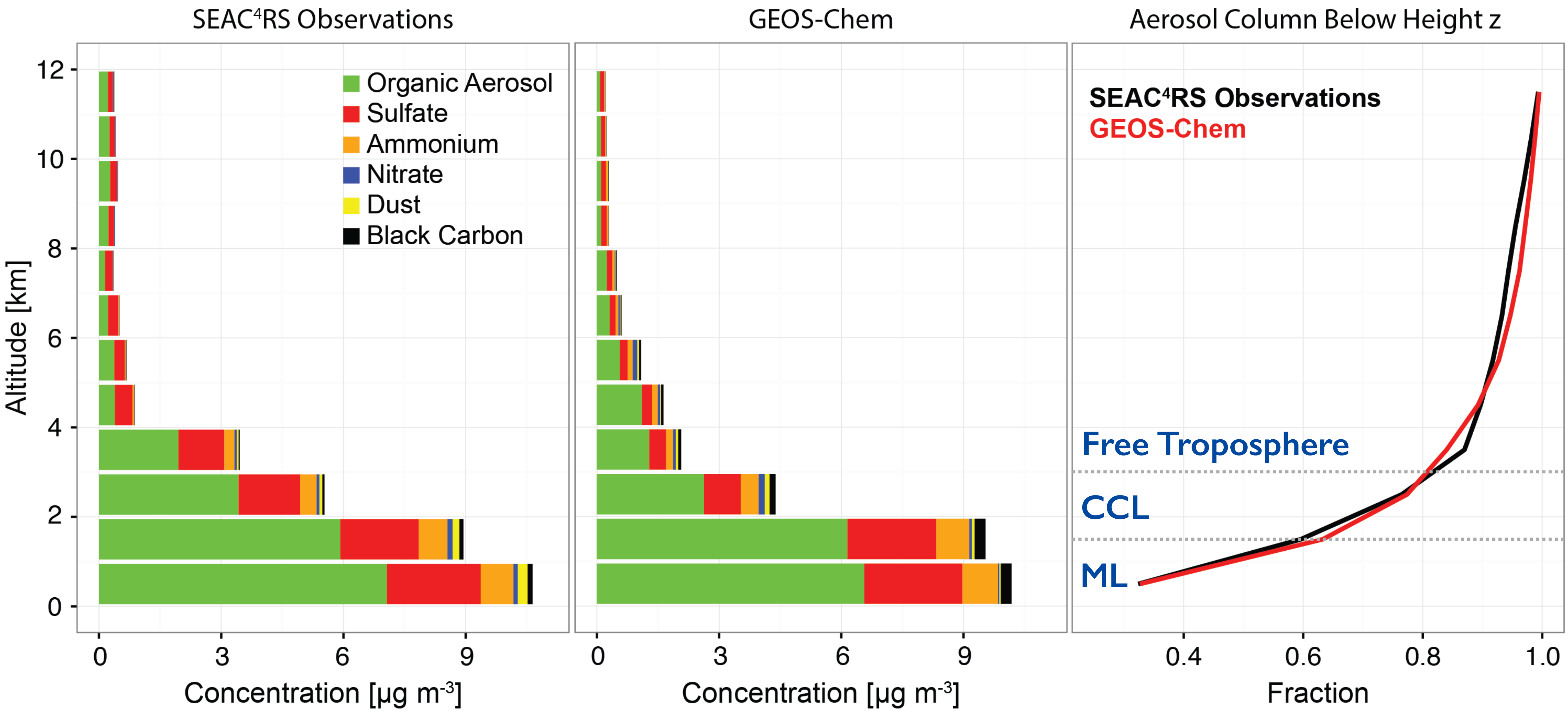
Aerosol Mass Drops Rapidly with Height



- Inconsistency between IMPROVE and AMS OA
- Little evidence for a large source of OA from aqueous phase processing in clouds
- Most aerosol ventilated from boundary layer rather than produced in free troposphere (fires and dust long-range transport are the exception)

AMS: Jose Jimenez, PALMS: Karl Froyd

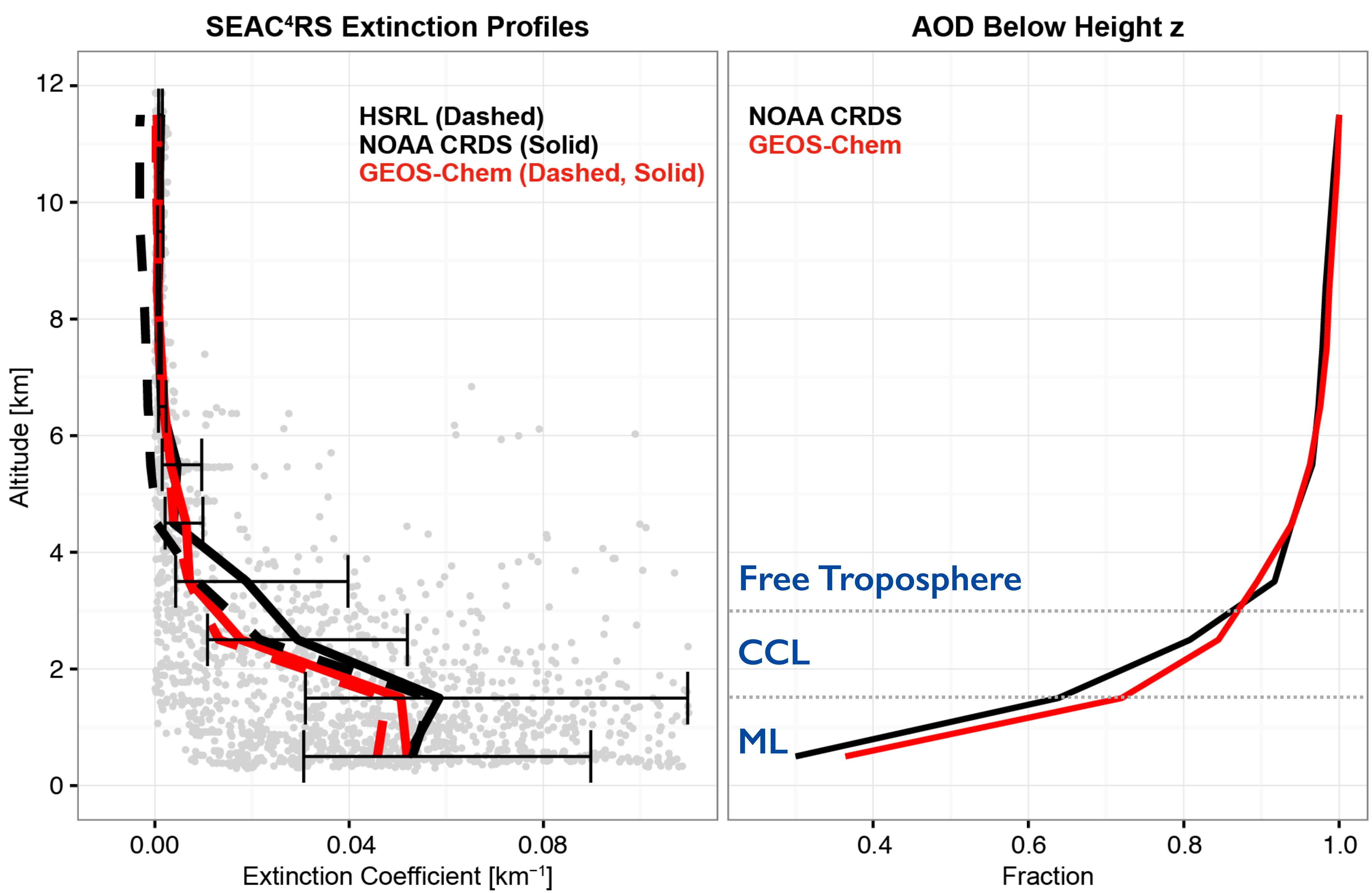
Organic Aerosol is Dominant Component at all Altitudes



- 40% of aerosol burden above the mixed layer (ML), 20% above the cloud convective layer (CCL)
- GEOS-Chem closely reproduces vertical profile of total aerosol mass (6% column underestimate)

AMS: Jose Jimenez, PALMS: Karl Froyd, SP2: Shuka Schwarz

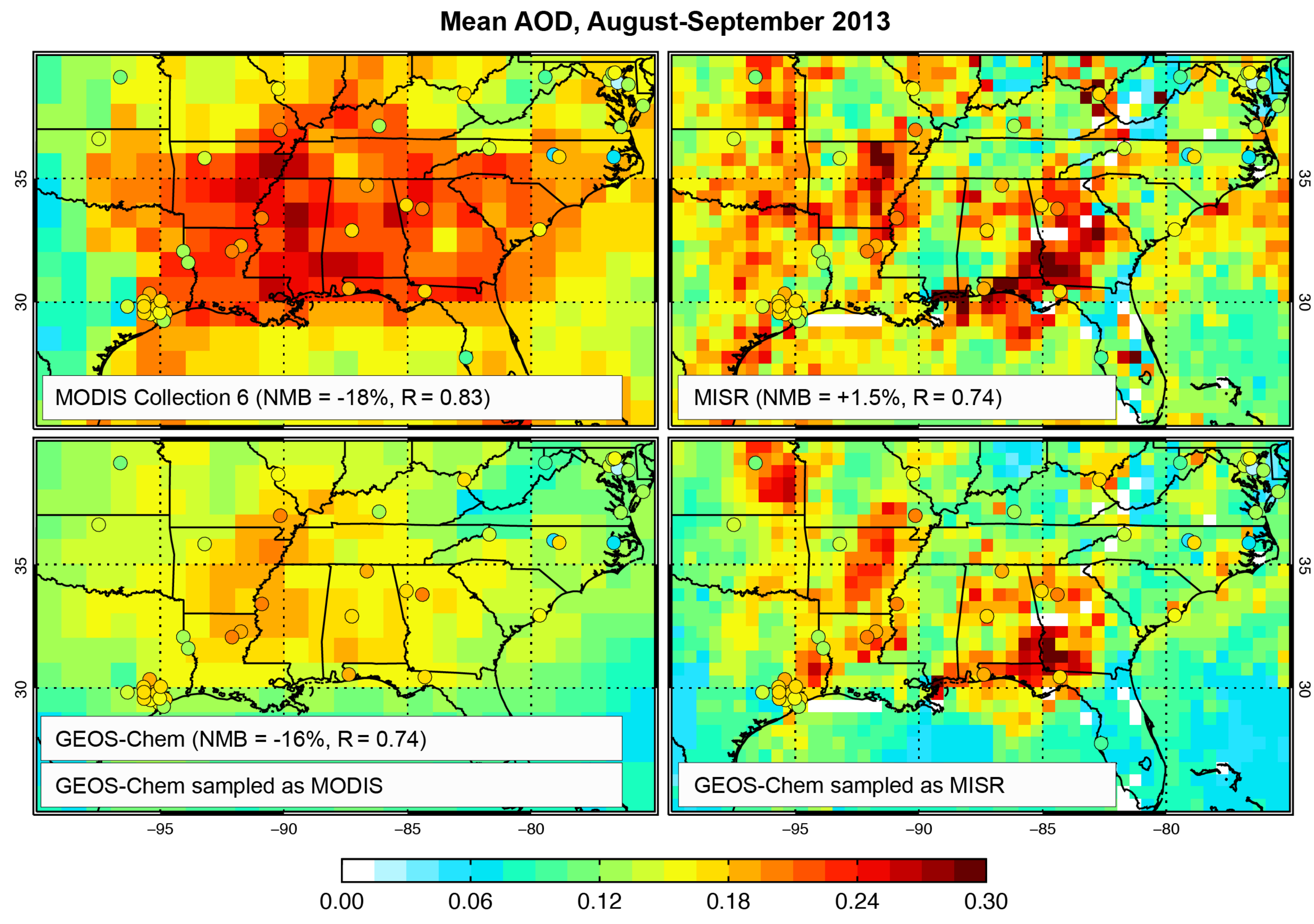
Extinction Profile Closely Follows that of Aerosol Mass



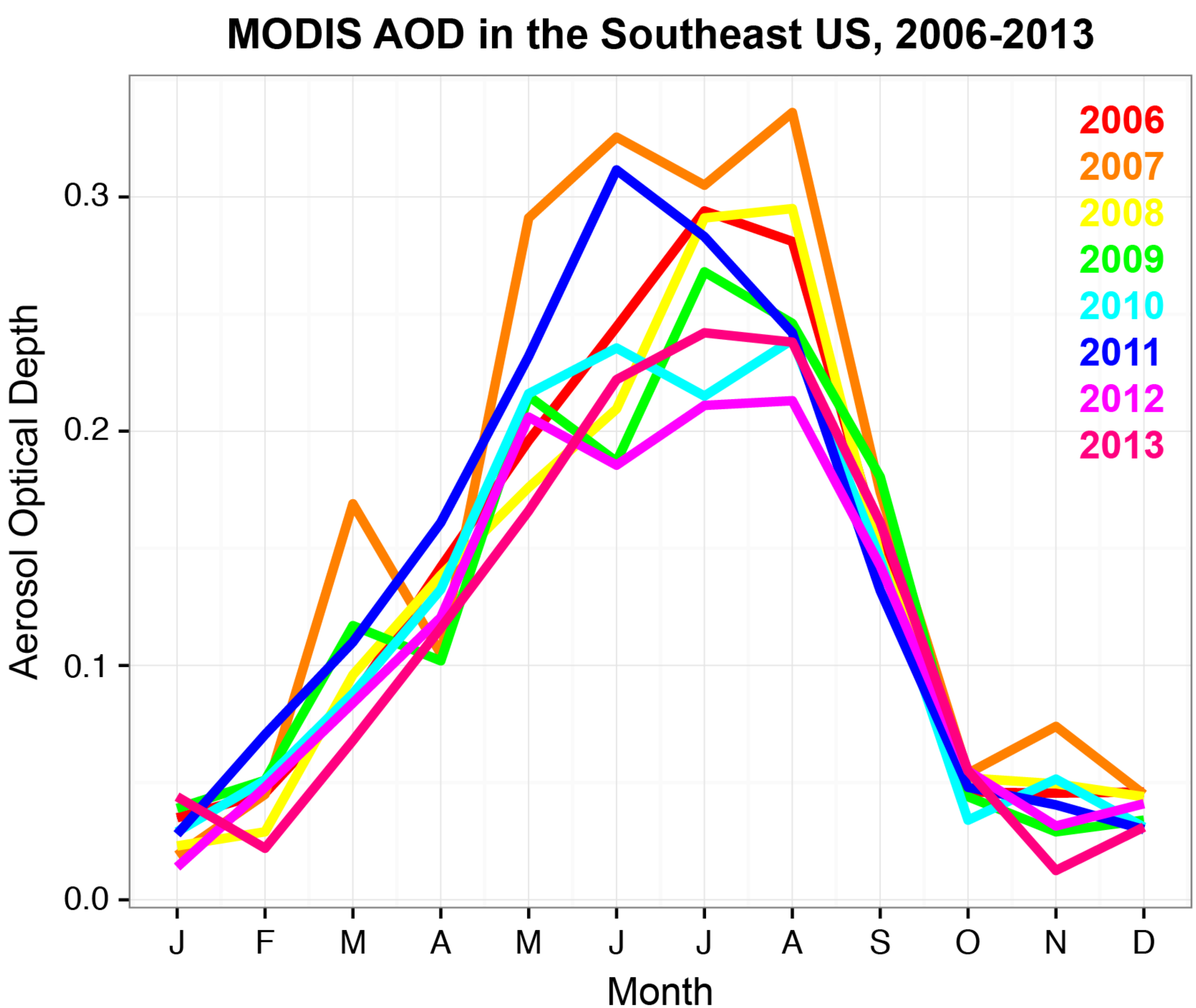
- Good agreement between two independent measurements
- GEOS-Chem underestimates column extinction by 16%

HSRL: Hair, CRDS: Wagner

Satellite AOD is Consistent with Surface $PM_{2.5}$

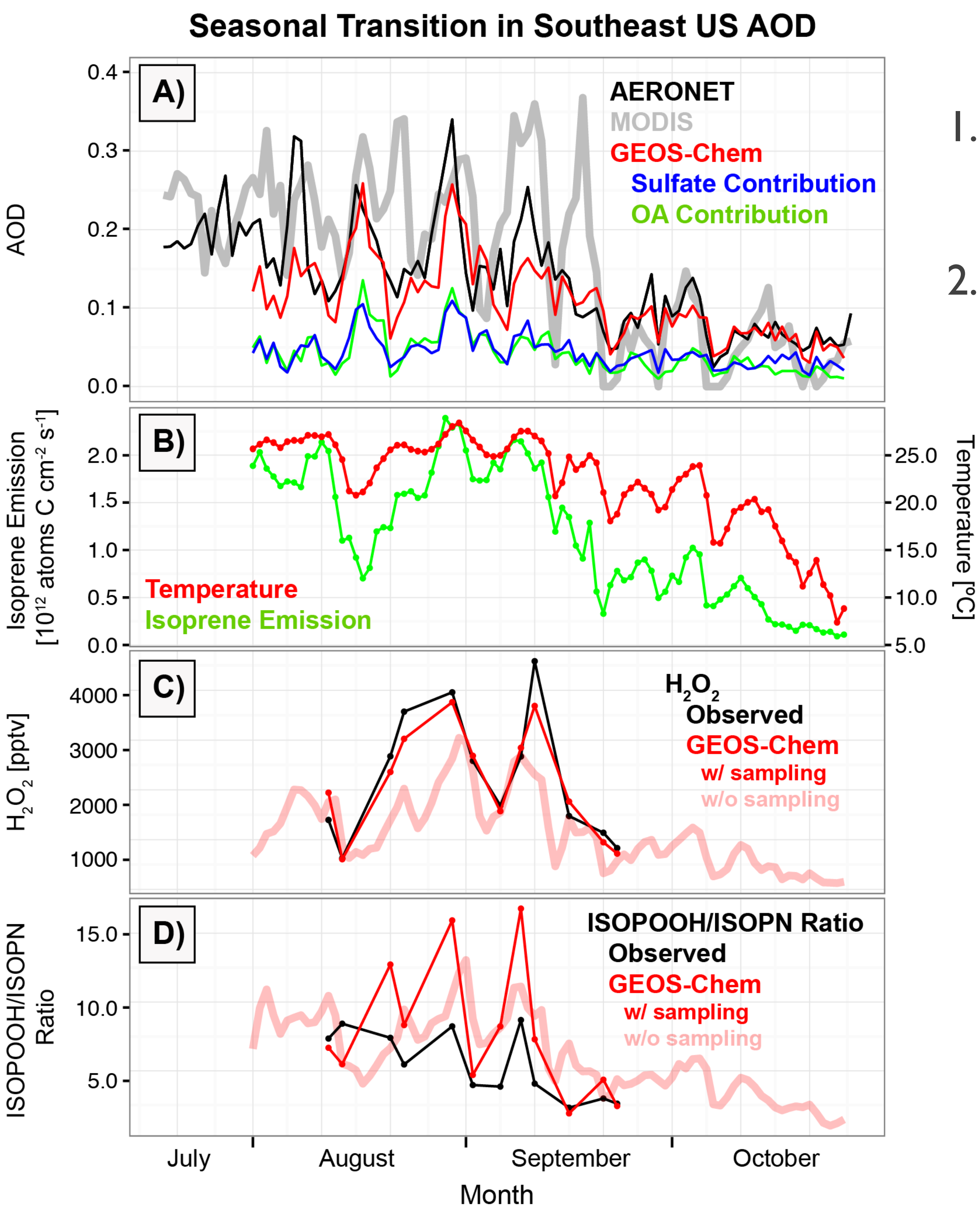


Aerosol Seasonal Cycle



- AOD seasonal cycle is driven by a sharp August - October transition in all years
- General decreasing trend in summer is interspersed by high fire years (2007, 2011, 2013)

The Seasonal Transition is Driven by Two Factors



1. Decline in biogenic emissions limits formation of OA and possibly sulfate
2. Decline in UV radiation causes a dramatic decline in H_2O_2 production

Corresponding shift in SOA formation with a transition from low NO to high NO conditions

CIMS: Wennberg

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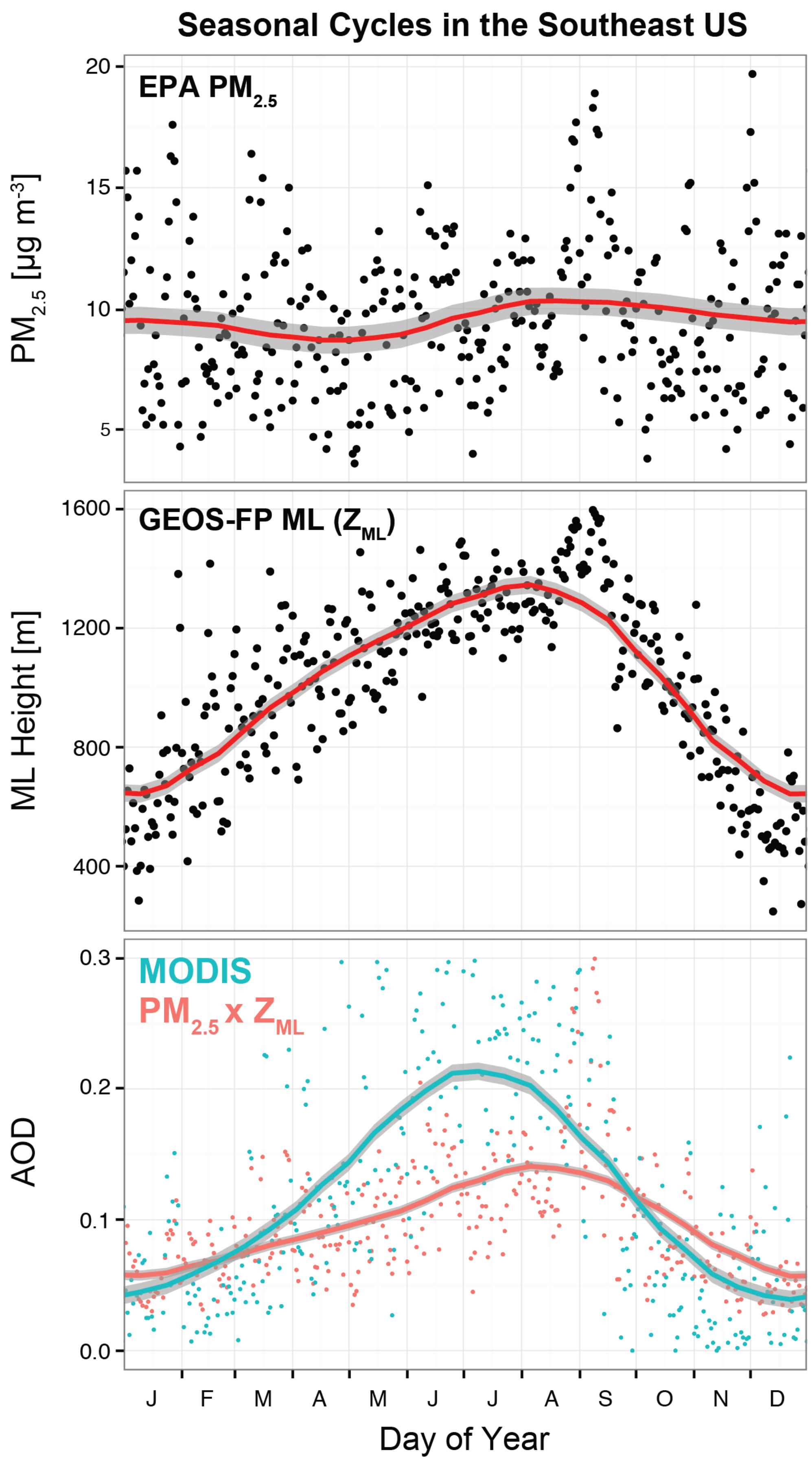

1. Decline in biogenic emissions limits formation of OA and possibly sulfate

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   Corresponding shift in SOA formation with a transition from low NO to high NO conditions

- Seasonality in mixed layer height and ventilation (25% extinction in CCL during SEAC^{4}RS) largely explains the difference between the seasonal amplitude of surface PM and satellite AOD

CIMS: Wennberg

Conclusions

- We find good consistency between surface, aircraft, and satellite observations of aerosol mass and light extinction, with the exception of IMPROVE OC measurements. Satellite measurements can be reliably used to infer $PM_{2.5}$ if a good CTM representation of PBL mixing and ventilation are available

- The successful simulation of sulfate in GEOS-Chem requires a missing oxidant that could be Criegee Intermediates, aqueous aerosols, or clouds

- OA is the dominant aerosol component in the Southeast US and is formed primarily from biogenic sources (40% isoprene, 20% monoterpenes). Successful simulation of the OA vertical profile argues against a large source in the free troposphere other than ventilation from the boundary layer

- The surface ammonium trend in 2003-2013 cannot be explained by a trend in emissions and is inconsistent with an extent of neutralization < 1. This inconsistency could be explained if their is inhibition of ammonia uptake by organic particle material

- The strong aerosol seasonal cycle in the Southeast US is related to a temperature-driven reduction in natural emissions and a rapid decline in UV radiation

- The apparent inconsistency between the magnitude of the surface $PM_{2.5}$ and satellite AOD seasonal cycles can be explained by the seasonal variation in mixed layer height and ventilation to the CCL